1   **Safe use of Electric Arc Furnace Dust (EAFD) as secondary raw**

2   **material in self-compacting mortars production**

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19   **Abstract**

20   This research contributes to the development of the Circular Economy concept by managing

21   waste through its use as a construction material. A novel process in which two samples of

industrial waste Electric Arc Furnace Dust (EAFD) collected from two different steelwork plants are used as secondary raw material (SRM) in the production of self-compacting mortars is proposed. At a previous stage, a characterisation of mortar material components was carried out. Then, self-compacting mortars with replacement ratios of 25%, 50% and 100% of siliceous filler with EAFD were designed. The feasibility of EAFD use was analysed by means of the study of mechanical strength, mineralogical, capillary properties and leaching behaviour in monolithic and granular state. The environmental classification of mortars, according to their leaching behaviour, was carried out in accordance with European Directive 2003/33/EC (2003). EAFD incorporation modified the self-compactability and common hydration reactions. Mechanical strength decreased with EAFD incorporation, although the mortar with 25% of replacement with one of the EAFD's showed a negligible loss compared to the self-compacting reference mortar. Water absorption by capillarity increased with EAFD content, consistently with the mortars' mechanical behaviour. The leaching behaviour analysis in a monolithic state revealed that all mortar releases were below the permitted limit. In a granular state, the mortar with the best mechanical behaviour was environmentally safe, leaving all the elements encapsulated by the self-compacting matrix. The results of this study contribute to EAFD management through its valorisation as secondary raw material in the production of self-compacting mortars, addressing an unprecedented line of research.

1. Introduction

The Circular Economy concept describes a cyclical system in which economic and environmental aspects are integrated, with the objective of reducing both raw materials consumption

46   and generated waste. The Circular Economy Implementation Plan of the European Parlia-

47   ment Commission (COM(2017)33, 2017) has highlighted the special importance of waste

48   management and of the use of secondary raw materials (SRM). Therefore, the recovery of

49   waste through its use as SRM is an attractive alternative in the fight against the scarcity of

50   natural resources.

51   The construction sector, specifically the concrete industry, stands out for its special role

52   within the Circular Economy, since this material is one of the most used and consumes

53   many resources (Rattanashotinunt et al., 2018). At a European level, the construction sec-

54   tor is considered one of the main sources of waste. Due to this, the use of waste in concrete

55   manufacturing is a goal that has been pursued for decades. The wastes traditionally used

56   are fly ash, electric arc furnace slag (Anastasiou et al., 2014; Kim et al., 2018) and con-

57   struction and demolition waste (López-Uceda et al., 2016; Silva et al., 2016), generating

58   beneficial situations in this industry.

59   Additionally, self-compacting concrete technology (SCC) has been one of the most im-

60   portant advances in construction materials. These fluid mixes can be placed and com-

61   pacted under their own weight, without vibration, segregation, blockage of coarse aggre-

62   gate, bleeding, or paste exudation (EFNARC, 2005). Because their use has been wide-

63   spread in recent years in civil engineering works, buildings and the precast industry, al-

64   ternative materials rather than conventional ones have gained relevance in concrete pro-

65   duction. In accordance with this, a wide range of wastes, such as filler of bituminous

66   mixes (Esquinas et al., 2017), filler from marble and tile waste (Tennich et al., 2015) and

67   electric arc furnace slag (Santamaría et al., 2017), has been investigated successfully, ob-

68   taining more environmentally-friendly self-compacting concrete.

69   During steel production by scrap melting as a raw material, metals such as Zn, Pb, Fe or

Cd are volatilised to later react with oxygen condensing into solid particles (Issa et al., 2012). These particles are named Electric Arc Furnace Dust (EAFD). Due to its high content of potentially leachable heavy metals, several directives such as the European Waste Catalogue (EWC, 2002) and the United States Environmental Protection Agency (USEPA, 2018) classify it as hazardous waste. The production of EAFD is estimated at around 15-25 kg per ton of steel produced (Sayadi and Hesami, 2017). Between 5 and 7 million tons of EAFD are produced worldwide (Kelebek et al., 2004).

Directly recycling EAFD in the furnace is difficult because the recirculation of volatile elements increases production costs and produces an accumulation of impurities in the melted steel mass (Nyirenda, 1991). In addition, EAFD is made of extremely fine particles (0.1 - 200 μm) (Lenz and Martins, 2007) and its handling is difficult without previous pelleting.

Globally, 70% of EAFD is landfilled, after treatment (Maslehuddin et al., 2011). Stabilization/Solidification (S/S) techniques are used in waste encapsulation, particularly for those containing heavy metals (Chen et al., 2009). Therefore, S/S is the technique generally used in the management of EAFD. The effectiveness of this process is studied by physical, chemical and microstructural methods, analysing the efficiency degree mainly through two parameters: mechanical strengths and leaching behaviour of the solids generated (Ledesma et al., 2017).

Although studies about S/S of EAFD have been successful (Laforest and Duchesne, 2006; Salihoglu and Pinarli, 2008), landfilling is not an option compatible with the current paradigm of Circular Economy, due to the use of natural resources during the treatment, such as natural aggregates or cement, and the content of valuable metals lost in this process.

In the search for alternative uses of EAFD to landfill, Sayadi and Hesami (2017) evaluated its effectiveness as a filler in asphalt mixes and Cholake et al. (2018) combined blue

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94 slate aggregate and EAFD in the production of high-strength lightweight aggregate for
95 use as construction material. Other authors, such as da Silva Magalhães et al. (2017),
96 report that the utilisation of EAFD in the construction industry is an interesting option
97 compared to landfilling, as it allows encapsulating hazardous elements in a stable matrix.

98 According to self-compactability properties, SCC is denser than conventional concrete,
99 which is why, this matrix could allow the encapsulation of hazardous waste type EAFD.
100 The need to have a greater volume of ultrafine material in the SCC could be combined
101 with the benefit of contributing to the elimination of the EAFD. This alternative has not
102 been investigated until now.

103 The aggregate content in SCC is lower than that of conventional concrete; so, part of the
104 performance of SCC is defined by the behaviour of the self-compacting mortar (SCM)
105 phase, one of the main components of SCC (Khotbehsara et al., 2018). Previous evalua-
106 tion of the mortar phase to analyse the characteristics of SCC has been carried out by
107 different authors, such as Nepomuceno et al. (2012) or da Silva et al. (2015).

108 Our study analyses the influence of incorporating two EAFD wastes as filler replacement
109 on the performance of SCM, evaluating the safety of their use through the mechanical
110 and leaching behaviour. This research has as objective to develop economic and sustain-
111 able techniques in construction and to search viable EAFD management, reducing the
112 consumption of natural resources for its treatment and avoiding deposit in hazardous
113 waste landfills. To the best of the authors' knowledge, this is the first research that pro-
114 duced SCM mechanically and environmentally safe, achieving EAFD/CEM ratios higher
115 than those available up to now. This line of research has not been previously explored
116 and promotes the development of the new Circular Economy paradigm.

2. Experimental details

2.1. Material characterisation

The materials used in the manufacturing of SCM were: two EAFD wastes, Portland cement, siliceous filler, two fractions of siliceous natural sand and a superplasticiser.

The wastes, named EAFD1 and EAFD2, came from two steelworks located in Olaberría and Zumárraga (Guipúzcoa, Spain). Both steelworks use an Electric Arc Furnace (EAF) for steel production. The EAFD samples were collected according to UNE-EN 14899:2007 (AENOR, 2017) from the electrostatic precipitators of the Electric Arc Furnaces. The cement used was CEMI/42.5R (CEM), supplied by SECIL Company (Lisbon, Portugal). The siliceous filler (SF) was provided by Minas Carmina Palau Saverdera (Gerona, Spain). Two fractions of aggregates were used in the mortars manufacture, fine sand 0/2 and coarse sand 0/4 (NS-0/2 and NS-0/4). The MasterEase 5025 (S_p) admixture was supplied by BASF Company. This is a superplasticiser/high-activity reducer of water for the production of high-strength, low viscosity and improved rheology concrete. The S_p structure is based on PAE polymers (polyacrylic ester) and has some analogy with that of polycarboxylates, but has important differences in the composition of the main chain.

The chemical composition of EAFD and NS was carried out by X-ray fluorescence (XRF) using 4 kW of power and S4PIONEER, BRUKER equipment. The CEM and SF composition was supplied by their manufacturers.

To obtain the X-ray diffraction (XRD) patterns of both EAFD, a device named Bruker D8 Discover A25 with Cu-Kα radiation was used. The goniometric scan was studied from 10° to 70° (2θ°) at a speed of 0.00625° min^{-1}.

Both EAFD were analysed by Fourier transform infrared (FTIR), by Bruker Tensor 27-Hyperion 2000 spectrophotometer (Bruker Optik GmbH, GERMANY) with CsI beam

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141   splitters and a DTGS detector. The transmission spectrum was collected using OPUSv

142   6.5 software.

143   The specific surface area of both EAFD and SF was analysed by the Brunauer-Emmett-

144   Teller method (BET), using Micromeritics ASAP 2010 equipment. The EAFD specific

145   gravity was determined by UNE 80103:2013 (AENOR, 2017). Regarding NS-0/2 and

146   NS-0/4, the specific gravity and water absorption was calculated according to UNE-EN

147   1097-6:2014 (AENOR, 2017). The specific surface area of CEM and specific gravity of

148   CEM and SF were provided by the manufacturers.

149   The particle size distribution of EAFD, CEM and SF was measured using a MastersizerS

150   device (Malvern Instruments) with ethanol as a dispersant of the samples. The particle size

151   distribution of NS-0/2 and NS-0/4 was tested in accordance with UNE-EN 933-1:2012

152   (AENOR, 2017). In addition, electron microscopy (SEM) was performed on both EAFD.

153   This technique was carried out by a scanning electron microscope JEOL JSM-6300.

154   EAFD1 and EAFD2 were analysed by means of the compliance test according to UNE-

155   EN 12457-4:2003 (AENOR, 2017) to study their leaching behaviour on the levels of

156   heavy metal release, listed in the European Directive 2003/33/CE (2003).

157   After the compliance tests, the eluates were analysed by Mass Spectrometry with Induc-

158   tively Coupled Plasma (ICP-MS) using a Perkin Elmer ELAN DRC-e spectrometer. The

159   leaching results obtained for the compliance test were compared with the legal limits es-

160   tablished by European Directive 2003/33/CE (2003) on the acceptance of landfill waste,

161   attached in Table 1, which allows environmental classification of materials based on the

162   release of their components.

163   **2.2. Mortar mix proportions**

164   A total of 7 mixes were manufactured in this study. All SCM were designed using the

165   Nepomuceno method (2005) according to the following criteria:

166   -   A value of 0.70 was chosen for the volumetric ratio (V_p/V_s), in absolute volume,

167   in all mortars. This ratio was the total amount of powder materials (V_p) and fine

168   aggregates in the mix (V_s). Nepomuceno et al. (2005) proposed volumetric ratio

169   values between 0.60 and 0.80;

170   -   With the aim of using EAFD as SF replacement, a reference mortar (SCRM) was

171   designed with 50% of CEM and 50% of SF. The mortars with waste proportions

172   were produced by replacing SF by EAFD in volume percentages (f_{Ad}) of 25%,

173   50% and 100%. Mortars with EAFD were named SCM-EAFDi-j:

174   o   i = EAFD type (EAFD1 or EAFD2);

175   o   j = the replacement percentage of SF by EAFD;

176   -   In this study, self-compactability was controlled via the relative spread area (G_m)

177   using the slump-flow test, and iteratively adjusting the V_w/V_p and $S_p/p_\%$ ratios.

178   This methodology has been explained in several previous works (Nepomuceno,

179   2005; Nepomuceno et al., 2012; Silva et al., 2011). V_w/V_p was the ratio in absolute

180   volume between the water and the powder materials and $S_p/p_\%$ was the ratio in

181   percentage between the amount in mass of superplasticiser (S_p) and powder ma-

182   terials (p). G_m was calculated according to equation (1) proposed by Nepomuceno

183   et al. (2012) and EFNARC (2005).

184   - $G_m = \left(\frac{Dm}{D_0}\right)^2 - 1$ (1)

185   -   Where D_m is the slump-flow average of the diameters recorded during the test and

186   D_0 the initial diameter of the base of the cone used in the test. D_m was kept in the

187   range of 270 ± 30 mm according to the values proposed by Kanadasan et al.

188   (2018), which was established as suitable for SCM.

189   Table 2 shows the composition of the mixes, the nomenclature used in this research and

190 the parameters applied to achieve self-compactability of the mortars. In addition, Table 3

191 shows the amount of constituents used per m^3 of mortar.

192 All mortars were mixed by the same procedure using a standard mixer (AENOR, 2017).

193 The constituents were homogenised in solid state for 30 seconds. Afterwards, 80% of

194 water was added. This paste was mixed for 2.5 minutes. Then, the remaining 20% of

195 water was added with the Sp diluted and mixing continued for 1 minute. All these steps

196 occurred at low velocity (140 rpm). After that, it was stopped for 1 minute and finally it

197 continued for 1 minute at a higher velocity (285 rpm).

198 From each mix, a total of 12 prismatic specimens (40 mm x 40 mm x 160 mm) were

199 made. The specimens were stored in their mould in a climatic chamber with 95% ± 5%

200 of relative humidity and 20 ºC ± 2 ºC of temperature for 24 hours, after which they were

201 demoulded and cured under the same conditions until testing.

202 **2.3. Test methods**

203 The flexural and compressive strengths of the hardened mortar were studied in accord-

204 ance with UNE-EN 1015-11:2000 (AENOR, 2017) at 7, 28 and 91 days using 3 speci-

205 mens per age.

206 XRD was used to analyse crystalline mineral components. For that purpose, hardened

207 mortars were crushed and sieved through a 0.125 mm sieve. The goniometric exploration

208 employed swept from 5° to 80° (2θ°) at a speed of 0.0142° min^{-1}.

209 The water absorption by capillarity was determined in mortars at atmospheric pressure

210 according to UNE-EN 1015-18: 2003 (AENOR, 2017) at 28 days of age. For this test 6

211 half specimens (40 mm x 40 mm x 80 mm) were used.

212 From an environmental point of view, the contaminating potential of EAFD must be an-

213 alysed in the production of SCM, guaranteeing their safe use. For this reason, leaching

214 tests were carried out, in monolithic and granular conditions, on the following mortars:

215 SCM-EAFD1-25, SCM-EAFD1-100, SCM-EAFD2-25 and SCM-EAFD2-100.

216 The specimens for leaching tests were protected from water and moisture with plastic

217 bags during the curing time so as not to affect the elements' mobility. The leaching tests

218 in monolithic and granular state were carried out in accordance with XP X31-211:2012

219 (AFNOR, 2017) and the compliance test UNE-EN 12457-4: 2003 (AENOR, 2017), re-

220 spectively, carrying out two repetitions of each test. To perform the compliance test, the

221 specimens were crushed and sieved previously. After the contact time of each test, an

222 eluate was analysed by ICP-MS.

223 Regarding the leaching limits of monolithic waste, Member States are responsible for

224 establishing the criteria to classify the material as inert, non-hazardous or hazardous.

225 However, while the monolithic European limits are established, the granular limits can

226 be applied, provided that the sample is crushed in advance. Due to the lack of European

227 monolithic limits, some European countries, such as Germany, Finland, Sweden or Den-

228 mark, apply the crushing of monolithic waste and its evaluation according to conventional

229 leaching tests. However, the English Environmental Agency (EEA, 2010) uses the limits

230 established in the European Directive 2003/33/CE (2003) for the monolithic waste veri-

231 fication without prior crushing.

232 In this study, all the leaching results obtained, in a monolithic and granular state, were

233 compared with the legal limits established by the European Directive 2003/33/CE (2003),

234 shown in Table 1.

3. Results and discussion

3.1. Materials characterisation

Table 4 shows the EAFD chemical composition results in oxides. The major components in both wastes were Zn and Fe at 38.27 and 29.88 for EAFD1 and 34.22 and 31.83 for EAFD2, respectively. The results agree with those of Vargas et al. (2006), López and López-Delgado (2002), Oustadakis et al. (2010) and Sapiña et al. (2014). The composition of these wastes strongly depended on the steel manufactured and a slight change in the furnace may lead to a different EAFD composition even within the same steelworks. The presence of heavy metals (Zn, Pb, Cr) affects the cement setting and jeopardize the mortar properties (Lasheras-Zubiate et al., 2011; Tashiro and Oba, 1979). Regardless of the EAFD composition, components must be able to be released in the environment, which can be determined by leaching tests. The pH value is a decisive factor (Ledesma et al., 2018). There are no previous studies of how heavy metals affect the self-compacting properties in mortars.

The chemical composition results of CEM, SF, NS-0/2 and NS-0/4, expressed in oxides content (%), are shown in Table 5. The main component of the CEM was calcium oxide (CaO) with lower contents of silica oxide (SiO_2), iron oxide (Fe_2O_3) or sulphur oxide (SO_3), while SF was 100% silica oxide (SiO_2). NS-0/2 and NS-0/4 had a siliceous character mainly with minority contents of aluminium oxide (Al_2O_3) and potassium oxide (K_2O) and iron oxide (Fe_2O_3).

Figure 1 shows the XRD patterns of both EAFD. The waste matrixes were composed mainly of Franklinite ($ZnFe_2O_4$) (22-1012) and Zincite (ZnO) (36-1451) (JCPD, 1995). These phases were also the main ones detected in the work carried out by da Silva

258   Magalhães et al. (2017). To a lesser extent, other phases were also detected, such as me-

259   tallic Mn (33-0887), manganese oxide (MnO) (07-0230), quartz (SiO_2) (33-1161) and

260   lead oxide (PbO_2) (37-0517) (JCPD, 1995).

261   The FTIR of EAFD1 and EAFD2 is shown in Figure 2. In these wastes, stretching and

262   bending vibration characteristic bands of the OH^- groups were observed (3504 and 1629

263   cm^{-1} in EAFD1 and 3643 and 1632 cm^{-1} in EAFD2, respectively) (Farmer, 1974). The

264   1170 and 1028 cm^{-1} bands were associated to asymmetric stretching vibration of Si-O-Si,

265   the 799 cm^{-1} band in EAFD1 to symmetric stretching vibration of Si-O-Si and the 453 or

266   427 cm^{-1} bands to bending vibration of O-Si-O. The 577 cm^{-1} band was associated to

267   bending vibration O-Al-O (Calvo et al., 2013; Farinha et al., 2018; Martins et al., 2008).

268   The presence of sulphates could be due to the 1087 cm^{-1} band (Nakamoto, 1986). In

269   EAFD1 and EAFD2 the presence of carbonates could be attributed to the 1425 cm^{-1} and

270   877 cm^{-1} bands, associated to asymmetric stretching vibration of CO_3^{2-} and bending out

271   of plane, respectively (Calvo et al., 2013). . It is usual that the FTIR technique detects the

272   presence of carbonate anion at low concentration levels, whose crystalline phases are not

273   detected by XRD. Table 4 shows the presence of calcium oxide, so it is not surprising the

274   presence of calcium carbonate, since calcium oxide hydrates and carbonates easily in the

275   presence of ambient humidity.

276   Table 6 shows the specific surface area, specific gravity and water absorption values for the

277   constituent materials of SCM. The specific surface area values of EAFD1 and EAFD2 were

278   3.70 m^2/g and 4.60 m^2/g, respectively. Law et al. (1983) reported that the EAFD specific

279   surface area was between 2.5 and 4 m^2/g using the BET method. The specific gravity was

280   3847 kg/m^3 for EAFD1 and 3809 kg/m^3 for EAFD2, in agreement with the results from de

281   Vargas et al. (2006). The specific surface area and specific gravity of CEM was 0.35 m^2/g

282 and 3140 kg/m^3, respectively. For SF, the specific surface area was 0.25 m^2/g and the spe-

283 cific gravity was 2600 kg/m^3. The specific gravity and water absorption of aggregates was

284 2537 kg/m^3 and 0.41% for NS-0/2 and 2622 kg/m^3 and 0.46% for NS-0/4.

285 Figure 3 shows the particle size distribution of EAFD, SF and CEM, in distribution per-

286 centage. CEM and SF exhibited a more homogeneous size distribution, whose values os-

287 cillate between 0.06 and 100 μm, with a majority size of 20 μm in both materials. EAFD1

288 and EAFD2 show bimodal curves with two major size fractions between 0.05-1 μm and

289 1-20 μm, with maximum located in 0.3 μm in both wastes and in 2.5 μm for EAFD2 and

290 in 3 μm for EAFD1. This distribution was in agreement with that reported by authors as

291 Oustadakis et al. (2010). These authors detected mainly two fractions of size: one fine

292 (0.1-1 μm) and one thicker (1-100 μm). Figure 4 shows the particles size distribution of

293 NS-0/2 and NS-0/4, in mass, where the maximum size was 2 and 4 mm for NS-0/2 and

294 NS-0/4, respectively.

295 Figures 5 and 6 show the EAFD SEM. Both figures show a typical spherical particle shape

296 of EAFD (Dutra et al., 2006) forming groups around 10-15 μm (Lenz and Martins, 2007).

297 Regarding leaching behaviour of both wastes, Figure 7 shows the concentration released,

298 expressed in mg/kg, for each element according to the compliance test UNE-EN 12457-

299 4: 2003 (AENOR, 2017). The parameters of conductivity (μS/cm), temperature (ºC) and

300 pH measured during the test were also reflected. Comparing the metal concentration val-

301 ues with the limit "Not hazardous" of European Directive 2003/33/CE (2003) of ac-

302 ceptance of waste in landfill (plotted in blue in Figure 7). It was observed that Se and Cd

303 exceeded the limit in EAFD1, while in EAFD2 it was Se, Mo and Pb. Due to this fact,

304 both wastes were classified as "Hazardous".

305 The Pb concentration in EAFD2 released into the medium was much higher (5483.87

306   mg/kg) than the Pb release in EAFD1 (6.14 m/g/kg). This phenomenon could be explained

307   by the higher pH of this EAFD2 (13.3) compared to the EAFD1 (9.6) registered during the

308   compliance test made according to UNE-EN 12457-4:2003 (AENOR, 2017). At a pH

309   higher than 12, Pb is mobilised easily (Pereira et al., 2001; Sebag et al., 2009; van der Sloot

310   and Dijkstra, 2004). Ledesma et al. (2018) studied the pH influence on the leaching of

311   EAFD. These authors revealed that the release of Pb was very low for pH values between

312   9 and 11. This justifies the low release of Pb in the EAFD1 and the high release in EAFD2.

313   **3.2. Self-compactability fresh parameters**

314   Table 7 shows the self-compactability parameters used in SCRM, SCM-EAFD1 and

315   SCM-EAFD2. In SCRM, the ratios V_w/V_p and $S_p/p_\%$ were established at 0.84 and 0.60 as

316   the optimal self-compactability parameters. The needs for water and S_p were higher when

317   EAFD was incorporated in the mixes. Properties such as fluidity, cohesion and the pass-

318   ing ability in SCM are very sensitive to changes in the free water content, the source of

319   the aggregate and the superplasticiser content (Kwan and Ling, 2017). For this reason,

320   the ratios V_w/V_p and $S_p/p_\%$ were modified with respect to SCRM to guarantee self-com-

321   pactability. EAFD was finer (Figure 3) and with greater specific surface (Table 6) waste

322   than SF, thus the water and S_p needs were higher than for SCRM.

323   Comparing the behaviour of mortars with EAFD1 and EAFD2, it is observed that the incor-

324   poration of EAFD1 requires a smaller amount of water (V_w/V_p) and superplasticiser ($S_p/p_\%$)

325   than EAFD2 to achieve the parameters of self compactability (Table 7). This trend could be

326   attributed to the particle size distribution of both wastes. In Figure 3, the largest volume of

327   particles enclosed between the curve and the range 0.05-1 μm corresponded to EAFD2

328   (50.92%, *versus* 41.52% in EAFD1). The greater number of fine particles in EAFD2 could

329   have caused a greater surface to hydrate. In addition, the greater specific surface area of

330   EAFD2 with respect to EAFD1 (Table 6) may have led to this behaviour.

331   Regarding G_m values, a decrease was observed as EAFD content increased, despite the

332   fact that the self-compactability parameters (V_w/V_p and $S_p/p_\%$) increased. The values of

333   G_m obtained, setting the ratios V_w/V_p and $S_p/p_\%$, were admissible due to none of the mixes

334   showing segregation of aggregate or water exudation visual signs.

3.3. Mechanical strength

336   The compressive and flexural strengths of mortars containing EAFD were evaluated at 7,

337   28 and 56 days of age according to UNE-EN 1015-11: 2000 (AENOR, 2017). The com-

338   pressive and flexural strengths results of SCM-EAFD1 are shown in Figures 8 and 9,

339   respectively. An abrupt loss of mechanical strength was observed with the incorporation

340   of EAFD1 at 7 days relative to SCRM. This loss cannot be attributed to the w/c ratio since

341   the variation was not significant when EAFD1 was incorporated (Table 7).

342   Figures 8 and 9 showed that SCM-EAFD1-25 increased mechanical strengths with values

343   of 0.98, 22.39 and 48.17 MPa for compressive strength and 0.15, 0.85 and 1.71 MPa for

344   flexural strength, at 7, 28 and 56 days, respectively. This phenomenon indicates a delay in

345   reaching mechanical strength relative to SCRM. Numerous studies have corroborated the

346   influence of heavy metals (Cu, Cr, Pb, Zn, Li, Hg) on the hydration and microstructure of

347   cement (Lasheras-Zubiate et al., 2011; Tashiro and Oba, 1979). Generally, the delay that

348   EAFD causes in the hydration of cement is attributed to the high concentration of Zn in the

349   composition of this waste (Table 4) (Balderas et al., 2001; Fares et al., 2016). Specifically,

350   the formation of a coating that envelops the cement particles, avoiding common hydration

351   (Yousuf et al., 1995). However, there are discrepancies about which Zn phase is responsible

352   for the coating. According to Yousuf et al. (1995), in solutions with sufficient presence of

353   Ca^{2+} ions, the different forms of zinc hydroxide ($Zn(OH)_3^-$ and $Zn(OH)_4^{2-}$) are transformed

354 into Calcium Zincate ($CaZn_2(OH)_6\cdot 2H_2O$), which completely covers cement grains, slow-

355 ing down the hydration reactions. However, Castellote et al. (2004), de Vargas et al. (2006)

356 and Brehm et al. (2017) state that it is Calcium Zincate ($CaZn_2(OH)_6\cdot 2H_2O$) that allows the

357 hydration of the paste to restart after the corresponding delay.

358 Castellote et al. (2004) observed a similar behaviour, where the reference mortar had a com-

359 pressive strength of 25 MPa and 38 MPa at 7 and 28 days, respectively. Mortars with an

360 EAFD/cement ratio equal to 0.86 did not gain compressive strength until 12 days, when it

361 was less than 3 MPa. However, at 28 days these mortars reached approximately 35 MPa. In

362 our study, SCM-EAFD1-25 has a lower EAFD/cement ratio (Table 3) than the one used by

363 Castellote et al. (2004) and compressive strengths lower than 35 MPa at 28 days.

364 The mechanical strength evolution of the mortars with 50% and 100% of EAFD1 was

365 different from that of those with 25%. The compressive and flexural strengths values rec-

366 orded by SCM-EAFD1-50 and SCM-EAFD1-100 were very similar at all ages of curing.

367 This phenomenon was related to the higher content of EAFD1 in mortars. The metal con-

368 tent in 50% and 100% replacement could cause the hydration reactions not to happen

369 before 56 days of curing. For SCM-EAFD1 mortars, a replacement percentage of SF with

370 EAFD1 greater than 25% is not viable from a mechanical point of view.

371 Although the compressive strength was considered insufficient for SCM-EAFD1-50 and

372 SCM-EAFD1-100, the values recorded at all ages exceeded the limits set by the English En-

373 vironmental Agency (EEA, 2010) and XP X31-211: 2012 (AFNOR, 2017) to be considered

374 as monolithic material deposited in landfill, although this is not the focus of the study.

375 Figures 10 and 11 show the mechanical strengths of SCM-EAFD2. In both figures an

376 adverse effect of EAFD2 was observed in the development of compressive and flexural

377 strengths, since these decrease with the higher EAFD2 content in mortars. This decrease

378 could be related to the following isolated and/or combined events: i) the higher EAFD2

379 content and consequently greater heavy metals presence in mortars that interfere in me-

380 chanical strength development; ii) the greater w/c ratio as a consequence of the greater

381 volume of fine particles and the high specific surface area of EAFD2.

382 Previous works by Ledesma et al. (2017) recorded lower compressive strengths compared

383 to the reference mortar, when the EAFD/cement ratio increased in cylindrical specimens of

384 conventional mortar made with limestone filler and aggregate. This behaviour was also

385 attributed to the greater presence of heavy metals that interfered in the common develop-

386 ment of mechanical strengths and greater need for water in mortars with EAFD. In the

387 research by da Silva Magalhães et al. (2017), the mechanical strength, with 5% and 10% of

388 EAFD as cement replacement, decreased with the increase in the w/c ratio.

389 All mortars with EAFD2 were viable from a mechanical point of view. Despite mechan-

390 ical strength reduction in SCM-EAFD2, the values of the SCM-EAFD2-25 mortar were

391 very close to those of SCRM at 56 days. These losses only represented 1.4%.

392 Mechanical strengths increased in all mortars as the curing age increased (mechanical

393 strength - $MS_{7d} < MS_{28d} < MS_{56d}$).

394 Figure 12 shows linear relationships of flexural and compressive strengths obtained by

395 the mortars at 7, 28 and 56 days, with good R^2 values. The flexural strength increases to

396 a lesser extent compared to the compressive strength at 56 days, which is consistent with

397 the generalized tendency for conventional concrete (Neville, 1995). This phenomenon

398 was also observed by da Silva Magalhães et al. (2017) when they evaluated the cementing

399 efficiency of EAFD in mortars.

3.4. Study of mineralogical phases

Figure 13 shows the mineral phases formed in SCRM during curing. The main detected phases correspond to Quartz (SiO_2) (33-1161) and Portlandite ($Ca(OH)_2$) (04-0733) (JCPD, 1995). The presence of Microcline ($KAlSi_3O_6$) (19-0932), Calcite ($CaCO_3$) (05-0586), dicalcium silicate (C_2S) (24-0034) and tricalcium silicate (C_3S) (42-0551) (JCPD, 1995) was also detected. At 56 days of curing, slight diffraction peaks associated to Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$) (41-1451) (JCPD, 1995) were also observed. This phase is formed during the cement hydration and, under normal curing conditions, it does not produce expansion (Skalny et al., 1996).

The X-ray diffraction patterns of hardened mortars containing EAFD are shown in Figures 14-19. As in SCRM, the prevailing phase was Quartz (SiO_2) (33-1161) (JCPD, 1995), due to the siliceous character of both the sands and filler used. Additionally, Franklinite ($ZnFe_2O_4$) (22-1012) and Zincite (ZnO) (36-1451) phases (JCPD, 1995), due to the composition of EAFD, were detected since diffraction peaks relative to these phases were detected in the wastes in their original state (Figure 1). A Cassiterite phase (SnO_2) (41-1445) (JCPD, 1995) was observed in the mortars containing EAFD1, due to the greater presence of Sn in their chemical composition. In mortars with 100% (Figures 16 and 19) and 50% (Figure 15) incorporation of EAFD1, Calcium Zincate ($CaZn_2(OH)_6 \cdot 2H_2O$) (25-1449) (JCPD, 1995) was detected, a characteristic phase of the reaction of mortar with EAFD (Castellote et al., 2004; de Vargas et al., 2006; Ledesma et al., 2017). Other phases, such as Microcline ($KAlSi_3O_6$) (19-0932), Orthoclase ($KAlSi_3O_6$) (31-0966), Calcite ($CaCO_3$) (05-0586), dicalcium silicate (C_2S) (24-0034) and tricalcium silicate (C_3S) (42-0551), Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$) (41-1451) and Iron sulphate hydroxide ($Fe_4(OH)_{10} SO_4/2Fe_2O_3 \cdot SO_3 \cdot 5H_2O$) (21-0429) (JCPD, 1995) were also detected.

Diffraction peaks associated with a Portlandite phase ($Ca(OH)_2$) (04-0733) (JCPD, 1995)

425   were observed in the SCM-EAFD1-25 mortars at 28 and 56 days (Figure 14). Additionally,

426   this phase also appeared in all mortars SCM-EAFD2 at 7, 28 and 56 days (Figures 17-19).

427   However, the Portlandite phase ($Ca(OH)_2$) (04-0733) (JCPD, 1995) was not detected in

428   SCM-EAFD1-25 at 7 days (Figure 14), neither in SCM-EAFD1-50 (Figure 15) nor in

429   SCM-EAFD1-100 at any age (Figure 16). Therefore, the mechanical strength achieved

430   could be related to the presence of Portlandite ($Ca(OH)_2$) (04-0733) (JCPD, 1995). The

431   amount of Portlandite ($Ca(OH)_2$) identified by DRX analysis is indicative of the extension

432   of the Portland cement reaction (Ledesma et al., 2017). The SCM-EAFD1-25 mortar's

433   XRD patterns (Figure 14) show that Portlandite ($Ca(OH)_2$) does not manifest itself after

434   7 days; however, it was detected at 28 and 56 days, due to the mechanical behaviour

435   previously explained. The addition of heavy metals, such as Cd, Ni or Pb to the paste

436   causes a decrease in the content of Portlandite ($Ca(OH)_2$), increasing their weakness

437   (Gollmann et al., 2010). In the XRD research of de Vargas et al. (2006), it was observed

438   that the content of Portlandite ($Ca(OH)_2$) decreases with the higher content of EAFD.

439   Figures 15 and 16 show the mineralogical phases of SCM-EAFD1-50 and SCM-EAFD1-

440   100, respectively. In these mortars, Portlandite ($Ca(OH)_2$) was not observed; but Calcium

441   Zincate ($CaZn_2(OH)_6 \cdot 2H_2O$) was. Ledesma et al. (2017) attributed the loss of mechanical

442   strengths of EAFD mortars EAFD to the formation of this phase ($CaZn_2(OH)_6 \cdot 2H_2O$) that

443   prevented the formation of Portlandite ($Ca(OH)_2$). In our research, the mechanical behaviour

444   of SCM-EAFD1-50 and SCM-EAFD1-100 mortars could be related to the appearance of

445   Calcium Zincate ($CaZn_2(OH)_6 \cdot 2H_2O$) as indicated by Ledesma et al. (2017). In mortars in

446   which Portlandite ($Ca(OH)_2$) was not detected, hydration may not have started and/or ended

447   because of EAFD presence. Conversely, regarding SCM-EAFD2-100, Figure 19 showed

448   joint formation of Portlandite ($Ca(OH)_2$) and Calcium Zincate ($CaZn_2(OH)_6 \cdot 2H_2O$), guaran-

449   teeing that both phases can be formed together during the hydration process, as stated by

450   Castellote et al. (2004), de Vargas et al. (2006) and Brehm et al. (2017).

451   Additionally (Figures 14-19), as the content of EAFD increases in the mortars, the content of

452   tricalcium silicate (C_3S) increases slightly for the same hydration age, indicating in turn a

453   lower presence of Portlandite ($Ca(OH)_2$) and worse mechanical behaviour. For the same mor-

454   tar with the same EAFD amount, it was observed that tricalcium silicate decreases (C_3S) and

455   Portlandite increases ($Ca(OH)_2$), with curing time, which will be the more pronounced the

456   lower the EAFD content. Balderas et al. (2001) carried out a study on tricalcium silicate (C_3S)

457   phase consumption in a cementitious paste doped with 10% EAFD. In this research, they

458   observed that the EAFD paste consumed less tricalcium silicate (C_3S) than the undoped mor-

459   tar, having a lower hydration reaction and worse final mechanical strength.

460   **3.5. Water absorption by capillarity**

461   The water absorption by capillarity results in accordance with UNE-EN 1015-18:2003

462   (AENOR, 2017) at 28 days are shown in Table 8. SCRM showed a water absorption value

463   by capillarity of 0.095 kg/($m^2 \cdot min^{0,5}$).

464   The water absorption by capillarity underwent a large increase when EAFD1 was incor-

465   porated into mortars, reaching a maximum value with SCM-EAFD1-50 of 1,077

466   kg/($m^2 \cdot min^{0,5}$). Regarding 50% and 100% replacement of SF by EAFD1, the mechanical

467   strength was not achieved, because of an incorrect and/or delay cement setting time, stop-

468   ping the formation of a cementitious matrix. High capillary coefficients represent high

469   absorption velocity values, which is undesirable from the point of view of mechanical

470   and leaching behaviour (Lucas et al., 2016). Therefore, the results of the SCM-EAFD1

471   mortars dissuade the use of a percentage higher than 25% replacement of SF.

472   In the SCM-EAFD2 mortars, capillarity increases as the EAFD2 content increased, ob-

473   taining a maximum value for the SCM-EAFD2-100 mortar with 0.209 kg/($m^2 \cdot min^{0,5}$).

474 Previous EAFD management studies have not taken this property into account. Figure 20

475 shows the relationship between capillarity and compressive strengths of mortars. It was

476 observed that mortars with better mechanical behaviour have less water absorption by

477 capillarity since there was a lower interconnection in the pores. This increase could be

478 related to the increase in the w/c ratio when incorporating EAFD2 (Table 7).

3.6. Leaching behaviour

480 The element release results according to XP X31-211: 2012 (AFNOR, 2017) of the mor-

481 tars SCM-EAFD1-25, SCM-EAFD1-100, SCM-EAFD2-25 and SCM-EAFD2-100 are

482 shown in Figures 21 and 22, respectively. In addition, the conductivity (µS/cm), temper-

483 ature (ºC) and pH recorded during the test are shown. The "Non-Hazardous" limit of the

484 European Waste Landfill Acceptance Directive (in blue) is also displayed. In mortars

485 SCM-EAFD1-25, SCM-EAFD1-100, SCM-EAFD2-25 and SCM-EAFD2-100, none of

486 the elements released into the environment exceeded the "Non-hazardous" limit estab-

487 lished by the European Directive (2003).

488 Ledesma et al. (2017) carried out a mechanical stabilisation study of EAFD using two

489 conventional mortars. In that study, mortar (cement and aggregate) was replaced by

490 EAFD with a ratio of EAFD/CEM of 3.33 and 6.67. Although the authors reduced the Pb

491 released in approximately 98%, this release exceeded the limit of the European Directive

492 (2003) and none of the mortars could classified as "Non-Hazardous". The authors at-

493 tributed the non-stabilisation of EAFD to the basic pH registered during the monolithic

494 leaching test. Our research succeeded in stabilising a maximum amount of 573.9 kg and

495 493.2 kg of EAFD/m^3 of mortar for EAFD1 and EAFD2, respectively, which is equiva-

496 lent to an EAFD/CEM ratio of 1.22. In our study, the effectiveness in monolithic state

497 resides in the higher density of the immobilization matrix. The SCM produced were

498 denser (values around 2.6 g/cm^3) than the conventional ones used by Ledesma et al.

499 (2017) (1.14 - 1.60 g/cm^3), allowing a better EAFD encapsulation.

500 Figures 23 and 24 show the elements released of the mortars SCM-EAFD1-25, SCM-

501 EAFD1-100, SCM-EAFD2-25 and SCM-EAFD2-100 in accordance with the compliance

502 test UNE EN-12457-4: 2003 (AENOR, 2017). These figures show the conductivity

503 (μS/cm), temperature (ºC) and pH measured during the test and the "Non-Hazardous"

504 limit established in the European Directive (2003) (coloured blue).

505 The SCM-EAFD1-25, SCM-EAFD1-100 and SCM-EAFD2-100 mortars exceeded the

506 "Non-Hazardous" limit of the European Directive (2003) due to the release of Pb with

507 values of 13,431, 40,521 and 44,301 mg/kg. It was observed that Pb release was higher

508 in mortars with greater content of EAFD.

509 The SCM-EAFD2-25 mortar was classified as "Non-Hazardous", because all elements

510 were kept below the legal limit in granular state, allowing a safe use of this material.

511 Several authors reported difficulty in incorporating Pb treatment into cementitious mate-

512 rials because the high alkalinity of cement makes its mobility easier (Ledesma et al., 2017;

513 Navarro et al., 2011). The pH of the cement-based materials ranges from 12 to 13. These

514 values coincide with the range of highest Pb release in the EAFD that is around 12

515 (Ledesma et al., 2018).

516 For 25% of replacement in both wastes, the Pb mobility could be more related to the phys-

517 ical characteristics of the matrix containing the waste. Figure 20 shows that mortars with

518 higher compressive strength have a lower water absorption by capillarity, therefore being

519 less accessible and susceptible to leaching. In this sense, the Pb in SCM-EAFD2-25 could

520 maintain its leaching (9,017 mg/kg) lower than the allowed limit (< 10 mg/kg) due to the

521 encapsulation matrix generated by the SCM, although its leaching was favourable from a

chemical point of view. The mobility of Pb is promoted in the alkaline environment produced by cement (Laforest and Duchesne, 2007). This mortar has values of compression strength and water absorption by capillarity very close to those of SCRM (Figure 20). The SCM-EAFD1-25 mortar was located further to the left in Figure 20, indicating lower compression strength values and greater water absorption by capillarity, thus favouring a greater Pb mobility in this matrix.

Because of the mortars location in Figure 20, SCM-EAFD2-100 (44,301 mg/kg) should have less leaching than SCM-EAFD1-100 (40,521 mg/kg). However, when the percentage of replacement of SF with EAFD was 100%, Pb leaching seems to be promoted by registered pH values and the greater amount of waste. Navarro et al. (2011) reported that the pH optimum to Pb precipitation was approximately 10. In the SCM-EAFD1-100 mortar (Figure 23), the difference between the pH of EAFD1 in the original state (9.6) and that reached through the compliance test (12.7) in this mortar could be responsible for the mobility of Pb. For SCM-EAFD2-100 (Figure 24) there was no significant difference between the pH of EAFD2 in the original state (13.3) and that reached during the compliance test (13.1). Therefore, this pH value encourages the leaching of Pb.

4. Conclusions

This study shows the influence of the use of two samples of electric arc furnace dust (EAFD1 and EAFD2), as replacement of siliceous filler (SF), used in the production of SCM. The main conclusions were:

- Both EAFD showed similar characteristics, with predominance of Zn and Fe oxides and mineralogical phases Franklinite ($ZnFe_2O_4$) and Zincite (ZnO). The specific surface area of EAFD2 (4.60 m^2/g) was higher than that of EAFD1 (3.70 m^2/g). The particle size distribution of both EAFD showed two fractions of sizes

546   and fine particles were higher in EAFD2. Regarding leaching behaviour, the waste

547   was classified as "Hazardous" since elements such as Se and Cd exceeded the

548   limit allowed in EAFD1, and Se, Mo and Pb in EAFD2. pH differences were

549   found in EAFD1 (9.6) and EAFD2 (13.3). The higher pH value in EAFD2 pro-

550   moted the high release of Pb that this waste registered;

551   -   SCM production using EAFD as raw secondary material was possible. When in-

552   corporating EAFD in the mortars, it was necessary to increase the amounts of

553   water and superplasticiser, to guarantee the self-compacting properties, due to the

554   higher content of fine particles and the bigger specific surface area of these wastes

555   compared to SF;

556   -   The mechanical strength decreased for higher EAFD content. From a mechanical

557   point of view, more than 25% replacement of SF with EAFD1 is not

558   recommended. Despite a delay in the gain of mechanical strength, the mortar with

559   25% of EAFD1 showed a compressive strength of 48.17 MPa at 56 days. The

560   mortars with EAFD2 showed lower strength losses, reaching 65.28 MPa of

561   compressive strength at 56 days for 25% of EAFD2. This value was very close to

562   that of the reference mortar at the same age (66.20 MPa);

563   -   Characteristic phases of EAFD hardened mortars, such as Franklinite ($ZnFe_2O_4$),

564   Zincite (ZnO) and Calcium Zincate ($CaZn_2(OH)_6 \cdot 2H_2O$), were detected. The

565   mechanical strength was related to Portlandite ($Ca(OH)_2$) formation. This phase

566   was not detected during the mineralogical study in the mortars with 50% and

567   100% replacement of SF with EAFD1;

568   -   Water absorption by capillarity increased with greater presence of EAFD, giving

569   rise to self-compacting matrices with more interconnected pores, resulting in

570   worse mechanical strength and matrices more susceptible to leaching;

571   -   The monolithic leaching test showed that all mortars were environmentally safe

572   because the elements showed releases below the established limit (10 mg/kg).

573   This was related to the SCM matrix that allows better EAFD encapsulation;

574   -   Pb mobility in mortar with 25% of EAFD2 in the granular state was related to the

575   physical characteristics of the self-compacting matrix. This mortar, due to its better

576   mechanical strength and lower capillarity, showed lesser susceptibility to leaching

577   relative to that with 25% of EAFD1 and it was the only one that was environmentally

578   safe to use. In mortars with 100% of EAFD1 and EAFD2 replacement, Pb leaching

579   was promoted by the pH values registered in the original state of the EAFD1 and

580   EAFD2 and those reached during the compliance test in granular state.

581   The results indicate that the use of less than 25% of EAFD1 in SCM is possible if Pb

582   release can be controlled in the granular state, because it is viable from a mechanical

583   viewpoint. Regarding the mortar with 25% of EAFD2, the requirements on mechanical

584   strengths and monolithic and granular leaching behaviour were achieved. EAFD2 up to

585   25% is viable to produce SCM, above which mechanisms that control the release of Pb

586   are necessary in the granular state. Through this research, a hazardous waste was saftely

587   managed through its incorporation in a construction process, such as the manufacture of

588   SCM, thus promoting Circular Economy.

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